

1 TO WHOM IT MAY CONCERN:

2

3 BE IT KNOWN THAT I, DAVID W. WARREN, a
4 citizen of the United States of America, residing in
5 Glendale, in the County of Los Angeles, State of
6 California, have invented a new and useful improvement
7 in

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10 COMPACT ENDOTHERMIC CATALYTIC

11 REACTION APPARATUS

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1 **BACKGROUND OF THE INVENTION**

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3 This application is a continuation-in-part of
4 Serial No. 09/687,098 filed October 16, 2000.

5 This invention relates to the use of
6 endothermic catalytic reaction apparatus operable to
7 produce hydrogen-containing gases from hydrocarbon
8 feedstock.

9 Endothermic catalytic reaction apparatus,
10 for converting hydrocarbon feedstock to hydrogen-rich
11 gases, is well known in the art. Commercial
12 production of hydrogen is commonly achieved by a
13 process known as steam reforming, that involves the
14 endothermic reaction between a mixture of hydrocarbon
15 feedstock and steam passed through a catalyst filled
16 reactor tubing that is heated.

17 In commercial steam reformers for large-
18 scale production of hydrogen from hydrocarbon feeds,
19 endothermic heat is commonly supplied by the
20 combustion of carbonaceous fuel and oxidant in a
21 diffusion or turbulent flame burner that radiates to
22 the refractory walls of a combustion chamber, thereby
23 heating them to incandescence, and providing a radiant

1 source for heat transfer to a tubular reaction
2 chamber. Uniform radiation to the surfaces of the
3 tubular reaction chamber is essential since excessive
4 local overheating of the tube surface can result in
5 mechanical failure. In large-scale commercial steam
6 reformers, mal-distribution of heat within the furnace
7 chamber is minimized by providing large spacing
8 between the individual reactor tubes, the furnace
9 walls, and the burner flames. However, for small-
10 scale catalytic reaction apparatus that is uniquely
11 compact, such as for the production of hydrogen for
12 small fuel cell applications, special design features
13 are needed to prevent tube overheating.

14 U.S. Patent 4,692,306 to Minet and Warren
15 describes a compact reformer comprising an annular
16 reaction chamber concentrically disposed around an
17 internal burner chamber containing a vertically
18 disposed cylindrical radiant burner that uniformly
19 radiates in the radial direction. A uniform radiation
20 pattern to a concentrically disposed annular reaction
21 chamber that surrounds the radiant burner, is
22 provided, thereby avoiding the problems with flame
23 impingement and local overheating of tube surfaces

1 that are associated with the use of diffusion or
2 turbulent flame burners in compact reformer apparatus.

3 However, there are practical limitations
4 regarding the use of an annular reaction chamber for
5 small-scale reformers having hydrogen production rates
6 of less than about 1500 SCFH. It is well known that
7 the heat transfer coefficient of gaseous reactants
8 contained within an annular reaction chamber is
9 directly related to the velocity of the gaseous
10 reactants within the annular space. In order to limit
11 the reaction chamber wall temperature, the velocity of
12 gaseous reactants within the annular space must be
13 sufficiently high to absorb the radiant heat flux that
14 impinges on the reaction chamber tube walls. However,
15 for very small-scale reformers, this requires that the
16 width of the annular reaction chamber space be small.
17 It is common practice in the art to limit the maximum
18 diameter of the catalyst particles packed within an
19 annular space to less than 20 percent of the width of
20 the annular space in order to ensure that the catalyst
21 is evenly distributed within the reaction chamber and
22 to prevent gas channeling along the walls of the
23 reaction chamber. However, for an annulus having a

1 small width dimension, this requires use of catalyst
2 particles of particularly small diameters thereby
3 resulting in an undesirably high pressure drop through
4 the catalyst bed.

5 The benefits of a flameless radiant burner
6 for use in compact catalytic reaction apparatus of
7 annular reaction chamber geometry are known. For
8 small-scale reformer applications, a tubular reaction
9 chamber geometry is preferred over annular reaction
10 chamber geometry in order to simultaneously achieve
11 high heat transfer coefficients and low pressure drops
12 within the reaction chamber.

13 There is need for a compact endothermic
14 catalytic reaction apparatus as embodied in the
15 present invention to achieve the objects of compact
16 design, while avoiding the problems of flame
17 impingement, excessive reaction chamber wall
18 temperatures, and excessive reaction chamber pressure
19 drop by application of a tubular reaction chamber that
20 is heated by the radiant burner. The tubular
21 endothermic reaction chamber as disclosed herein
22 employs a combination of catalyst particle sizes and
23 reactant mass velocities to control the reactor

1 pressure drop and the maximum reaction chamber tube
2 wall temperature within certain needed limits; and the
3 radiant burner is operated at specific ranges of
4 combustion intensity and excess air to control surface
5 temperature of the radiant burner within certain
6 needed limits. The present invention extends the
7 practical range of tubular endothermic reaction
8 chamber geometry that can be used in combination with
9 radiant burners for converting hydrocarbon feedstock
10 to useful industrial gases.

12 SUMMARY OF THE INVENTION

14 It is the general object of this invention
15 to provide a novel endothermic catalytic reaction
16 apparatus for the production of industrial gases from
17 a hydrocarbon or methanol feedstock that is
18 simultaneously compact, thermally efficient, has
19 improved life expectancy and low pressure drop, and is
20 particularly well suited for the small scale
21 generation of useful gases for fuel cell applications
22 in the range of 1 k W to 50 k W.

1 In the present invention, a compact burner
2 chamber employing a radiant burner assembly is
3 configured to distribute radiant energy along the
4 axial length of a tubular reaction chamber. In one
5 embodiment, the radiant burner assembly comprises a
6 woven metal fiber attached to a support structure that
7 permits the efflux of fuel and oxidant from the burner
8 core to the outer surface of the metal fiber. The
9 properties of the metal fiber stabilize the combustion
10 in a shallow zone proximal to the outer surface of the
11 metal fiber. The combustion reaction heats the metal
12 fiber to incandescence and provides a source of
13 radiant energy that is transferred to the reaction
14 chamber. In another embodiment, the radiant burner
15 assembly comprises a porous ceramic fiber burner that
16 accomplishes the same object by serving as a radiant
17 source of energy.

18 The metal fiber of the burner typically
19 consists essentially of an alloy containing
20 principally iron, chromium, and aluminum and smaller
21 quantities of yttrium, silicon, and manganese having
22 extended life at operating temperatures up to 2000°F.

1 In one embodiment, the tubular reaction
2 chamber has U-shape, and is sometimes referred to as a
3 hairpin tube, which is substantially filled with
4 catalyst, the tube extending into and out of the
5 combustion chamber for gaseous flow through. The
6 radiant burner axis is preferably vertically disposed
7 within the combustion chamber and oriented parallel to
8 the axis or axes of the U-tube reaction chamber. The
9 active radiant surface of the cylindrical radiant
10 burner assembly is defined by a geometric arc that
11 bisects the cylindrical assembly so as to maximize the
12 flux of radiant energy that is directed to the surface
13 of the U-tube reaction chamber. In this embodiment,
14 the center to center spacing between the radiant
15 burner and the U-tube reaction chamber, and the
16 radiation angle of the radiant burner are
17 simultaneously controlled, or configured for high
18 efficiency of heat transfer.

19 In a third embodiment, the tubular reaction
20 chamber comprises a helical coil that is substantially
21 filled with catalyst and has inlet and outlet portions
22 that pass into and out of the combustion chamber. The
23 helical coil is wrapped to form turns at specific lead

1 angles, so that the coil free area is in the range of
2 50% to 75%, wherein the free area is defined by the
3 ratio of the free area between helical tube conduits
4 or turns and the cylindrical surface that bisects the
5 helical coil circle or cylinder. The radiant burner
6 axis is typically vertically disposed within the
7 combustion chamber and the cylindrical radiant burner
8 is located at the center of the helical coil. In this
9 embodiment, the active radiant surface of the
10 cylindrical radiant burner assembly is defined by a
11 360-degree arc.

12 In each embodiment, the radiant burner is
13 operated at a combustion intensity and an excess air
14 ratio that is carefully controlled to limit the
15 radiant burner surface temperature to less than 2000°F,
16 and preferably in the range of 1500°F to 1900°F, in
17 order to provide extended life for the radiant burner.

18 In each embodiment, the catalyst particle
19 diameters and reactant mass velocities are carefully
20 controlled to simultaneously limit the reactor
21 pressure drop to less than 8 psi, and preferably in
22 the range of 2 psi to 4 psi in order to limit the

1 delivery pressure required for the hydrocarbon feeds,
2 and to limit the reaction chamber tube wall
3 temperatures to less than 1600°F, and preferably in the
4 range of 1300°F to 1500°F, in order to allow extended
5 life of the tube using relatively inexpensive tube
6 alloys.

7 In each embodiment, a portion of the
8 combustion chamber is configured to form an annular
9 convective chamber to enhance heat transfer from the
10 combustion products to the tubular reaction chamber.

11 A further object is to provide endothermic
12 catalytic reaction apparatus, comprising

13 a) a combustion chamber,
14 b) a tubular reaction chamber having two
15 generally tubular legs extending in generally parallel,
16 spaced apart relation within the combustion chamber,

17 c) catalyst within said reaction chamber
18 for reacting with a hydrocarbon and steam received
19 within the reactor chamber, to produce hydrogen and
20 carbon dioxide,

21 d) a radiant burner within the combustion
22 chamber and extending in generally parallel relation to
23 at least one of said legs, said burner spaced from said
24 legs,

1 e) said two legs having axes, and said
2 burner having an axis which is spaced in offset
3 relation to a plane defined by said leg axes.

4 In yet another embodiment, the tubular
5 reaction chamber comprises a straight tubular outer
6 conduit concentrically disposed around an inner
7 conduit. Catalyst is contained in the annular space
8 between the outer conduit wall and the inner conduit
9 wall. The tubular reaction chamber is configured so
10 that the flow of reactant gas is directed
11 longitudinally through the annular catalyst space in
12 one direction and returns down the inner conduit space
13 in the opposite direction. A portion of the tubular
14 reaction chamber extends into the combustion chamber.
15 One end of the tubular reaction chamber, containing
16 both an inlet means that is in communication with the
17 annular catalyst space and an exit means that is in
18 communication with the inner conduit space, extends
19 outside of the combustion chamber. A radiant burner is
20 oriented to direct a flux of radiant energy to the
21 surface of the outer conduit of the tubular reaction
22 chamber. If a multiplicity of such tubular reaction
23 chambers are used, they can be oriented concentrically
24 around a centrally disposed radiant burner that
25 uniformly radiates in a 360 degree arc. The radiant

1 burner may consist of metal fiber material, or ceramic
2 fiber material.

3 These and other objects and advantages of
4 the invention, as well as the details of an
5 illustrative embodiment, will be more fully understood
6 from the following specification and drawings, in
7 which:

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9 **DRAWING DESCRIPTION**

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11 Fig. 1 is an elevation showing assembled
12 components of the endothermic catalytic reaction
13 apparatus;

14 Fig. 1a is a section taken on lines 1a-1a of
15 Fig. 1;

16 Fig. 2 is a diagrammatic view of dimensional
17 characteristics of the Fig. 1 and 1a assembly;

18 Fig. 3 is a view like Fig. 1, but showing a
19 modification;

20 Fig. 3a is a section taken on lines 3a-3a of
21 Fig. 3;

22 Fig. 4 is a view like Fig. 1, but showing an
23 additional modification; and

1 Fig. 5 shows another embodiment of the
2 reaction apparatus.

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4 **DETAILED DESCRIPTION**

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6 The catalytic reaction apparatus seen in Fig.
7 1 depicts a preferred embodiment of the present
8 invention. The apparatus comprises a combustion
9 chamber 4, a convection chamber 17 extending into
10 chamber 4, and a reaction chamber 16. The combustion
11 chamber 4 is defined by the zone enclosed or surrounded
12 by refractory insulation 6. The reaction chamber 16 is
13 defined by the volume enclosed by tubular reactor
14 conduit 1. The tubular reactor conduit 1 is formed in
15 a U-tube or hairpin configuration having parallel
16 upright legs 1a and 1b, and a U-shaped bend 1c, and can
17 be removed from the combustion chamber upon removal of
18 a top flange 18. Leg 1b of the tubular reactor conduit
19 1 passes concentrically through the convection chamber
20 17 defined by the space enclosed between the convection
21 conduit 10 and the leg 1b of the tubular reactor
22 conduit 1. The reaction chamber including 1a, 1b, and
23 1c is packed with catalyst from the inlet fitting or
24 means 2, where reactants enter, to the outlet port or

1 means 3 where products exit. Convection conduit opens
2 at 13 to chamber 4, and discharges at 11.

3 An axially extending, vertically disposed
4 radiant burner 7 is supported by a burner gas conduit
5 12 that conveys a mixture of fuel and oxidant from an
6 inlet means 8 to the radiant burner. In this
7 embodiment, the radiant burner 7 comprises a gas
8 permeable metal fiber zone 14 and a non-permeable zone
9 16. Fuel and oxidant pass through the permeable metal
10 fiber zone 14 where they are ignited on the surface
11 thereby combusting and releasing heat to form an
12 incandescent zone that radiates energy outward in an
13 arc 15. The arc angles γ_1 and γ_2 of 14 and 16 are
14 such (angle of 14 is between 45° and 180°) that the
15 radiating pattern maximizes the flux of radiant energy
16 to the surfaces of the tubular reactor legs 1a and 1b,
17 and also U-bend 1c, while minimizing the flux of
18 radiant energy to the internal wall 19 of combustion
19 chamber 4. Fuel and oxidant are initially ignited on
20 the surface of the permeable metal fiber zone 14 using
21 an igniter 9. Once ignited, the combustion reaction on
22 the surface of the metal fiber zone 14 facing 1a and 1b
23 is self-sustaining.

24 The radiant arc angle of 14 is selected so
25 that the direct radiant flux from the burner that
26 bisects the projected surface of the reaction chamber

1 tube wall is a minimum of 50% of the total radiation
2 flux that emanates from the active radiant burner
3 surface. As an illustration of the condition, Fig. 2
4 depicts a geometric representation of the preferred
5 embodiment of the present invention. The active
6 radiant zone 14 emits radiation along a line of sight
7 defined by a radiant arc 15 that impinges on the
8 reaction chamber conduit legs 1a and 1b and the inner
9 surface 19 of the combustion chamber. The emitted
10 radiation is bisected by hypothetical plane 50 passing
11 through the centerline of the U-tube reaction chamber.
12 The projected area of the reaction chamber surfaces per
13 unit tube length receiving direct radiation from the
14 burner within the controlled radiant arc is given by
15 $a + a = 2a$, where 'a' is the outer diameter of each
16 leg. The total radiation within the arc 15 is given by
17 $c + c + a + a + b = 2c + 2a + b$. The dimensions 'a',
18 'b' and 'c' are as shown. In the preferred
19 embodiment of the present invention, the ratio of 2a
20 divided by $2c + 2a + b$ is typically greater than 0.5 or
21 50%.

22 In the present invention, the radiant burner
23 combustion intensity is controlled in the range of
24 150,000 btu/ft²/h and 350,000 btu/ft²/h wherein the
25 combustion intensity is defined as the higher heating
26 value of the fuel combusted divided by the permeable

1 radiant burner surface area and the excess combustion
2 air operating air ratio is controlled in the range of
3 30% to 100% (wherein the excess air ratio is defined as
4 percent combustion air in excess of the stoichiometric
5 amount required for complete combustion of the burner
6 fuel) to prevent overheating of the surface of the
7 radiant burner and to prevent overheating of the
8 premixed fuel and oxidant contained within the burner
9 core. In the present invention, the reactant mass
10 velocity is controlled in the range of 400 lb/ft²/h to
11 1500 lb/ft²/h in order to limit the reaction chamber
12 tube wall temperature to the desired range of 1300°F to
13 1500°F.

14 Combustion products emanating from the
15 permeable metal fiber zone 14 enter the inlet 13
16 leading to the convection chamber 17, wherein the
17 combustion products exchange heat with tubular reaction
18 chamber 1 for preheating the feed to leg 1b.

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20 **EXAMPLE**

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22 A compact endothermic catalytic reaction
23 apparatus according to the preferred embodiment was
24 constructed and tested. The reaction chamber consisted
25 of 1 inch schedule 40 pipe constructed of 310 stainless

1 steel that was formed in a U-tube arrangement spaced on
2 3 inch centers. The reaction chamber was packed with a
3 commercial steam reforming catalyst that was crushed
4 and screened to an average particle size of
5 approximately $\frac{1}{4}$ inch.

6 The radiant burner consisted of 4 inch long
7 by 1 $\frac{1}{2}$ inch outer diameter cylindrical assembly that
8 had an active radiant angle γ_1 of 120 degrees. The
9 burner assembly was placed in an insulated combustion
10 chamber having dimensions of 6 inch internal diameter
11 and 10 inch height. The radiant burner assembly was
12 spaced approximately 4 inches from the U-tube
13 centerline. The convection chamber consisted of a 2
14 inch tube constructed of 304 stainless steel.

15 The radiant burner was fired using a mixture
16 of propane and air at a total higher heating value
17 firing rate of 12,000 btu/h. The reactant mixture
18 consisted of 1 lb/h of propane and approximately
19 3.5 lb/h of steam and was fed to the reaction chamber
20 at a temperature of approximately 800°F. The reactant
21 mixture was heated in the reaction chamber to an exit
22 temperature of 1250°F. The measured tube wall
23 temperature of the reaction chamber was 1450°F, the
24 radiant burner surface temperature was 1750°F, and the
25 combustion products exit temperature was 1050°F. The

1 estimated hydrogen plus carbon monoxide yield was 67
2 SCFH.

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4 Fig. 3 depicts another embodiment of the
5 present invention. In this embodiment, a radiant
6 burner surface 30 having a hemispherical geometry
7 radiates energy to the reaction chamber like that of
8 Fig. 1. A mixture of fuel and oxidant enters the
9 radiant burner from an inlet conduit 31. The
10 longitudinal axis of the inlet conduit is oriented
11 normal to the plane of the U-tube reaction chamber.

12 Fig. 4 depicts yet another embodiment of the
13 present invention. In this embodiment, the reaction
14 chamber is defined by a volume enclosed by a tubular
15 reactor conduit comprising an upper section 19
16 consisting of a vertically disposed tube that is
17 connected to the inlet means 2, a lower section 20
18 consisting of a helical coil, having an outer diameter
19 between 6 and 36 inches, and an exit section 21
20 consisting of a vertically disposed tube that is
21 connected to an exit means 3. The upper section 19 of
22 the tubular reactor conduit passes concentrically
23 through the convection chamber 17. The reaction
24 chamber is packed with catalyst from the inlet means 2,
25 where reactants enter, to the outlet zone 22 of the

1 lower section 20. The reaction chamber has outer
2 diameters ranging from $\frac{3}{4}$ inch to 4 inches.

3 An axially extending radiant burner 7 is
4 vertically disposed along the central axis of the
5 helical coil section 20 of the tubular reaction
6 conduit. The radiant burner is supported by a burner
7 gas conduit 12 that conveys a mixture of fuel and
8 oxidant from the inlet means 8 to the radiant burner.

9 In this embodiment, the radiant burner 7 comprises a
10 gas permeable metal fiber zone 14 that subtends the
11 entire circumference of the radiant burner. Fuel and
12 oxidant pass through the permeable metal fiber zone 14
13 where they are ignited on the surface, thereby
14 combusting and releasing heat to form an incandescent
15 zone that radiates energy in a predominantly uniform
16 radial direction. The helical tubular reaction chamber
17 and catalyst therein are sized for creation of mass
18 velocities ranging from 400 lb/ft²/h to 1500 lb/ft²/h.
19 The catalyst in the helical tubular reaction chamber
20 has average catalyst particle diameters ranging from $\frac{1}{4}$
21 to 1 inch for producing gas pressure drops ranging from
22 1 psi to 8 psi during flow through the reaction
23 chamber. The helical tubular reaction chamber has gas
24 exit end temperature ranging from 1150°F to 1400°F,
25 when heated by said radiant burner, in operation. The
26 helical tubular reaction chamber has maximum tube wall

1 temperatures ranging from 1300°F to 1600°F, when heated
 2 by said radiant burner, in operation. The helical
 3 tubular reaction chamber has average heat fluxes
 4 ranging from 3,000 btu/ft²/h to 10,000 btu/ft²/h, when
 5 heated by said radiant burner in operation. The
 6 helical tubular reaction chamber is sized to have
 7 capacity to generate hydrogen plus carbon monoxide
 8 product in volumetric quantities ranging from 50 SCFH
 9 to between 100 and 1500 SCFH. The radiant burner
 10 comprises a supported metal fiber material consisting
 11 essentially of an alloy containing principally iron,
 12 chromium, and aluminum and smaller quantities of
 13 yttrium, silicon, and manganese, said alloy having
 14 extended life at operating temperatures up to 2000°F.
 15 The radiant burner has surface temperatures ranging
 16 between 1500°F and 1900°F, in operation. The radiant
 17 burner has an operating combustion intensity typically
 18 ranging from 150,000 btu/ft²/h to 350,000 btu/ft²/hr,
 19 wherein the combustion intensity is defined as the
 20 higher heating value of the fuel combusted divided by
 21 the permeable radiant burner surface area. The radiant
 22 burner has an operating excess air ratio typically
 23 ranging from 30% to 100%, wherein the excess air ratio
 24 is defined as percent combustion air in excess of the
 25 stoichiometric amount required for complete combustion
 26 of the burner fuel. The helical coil has free area in

1 the range 50% to 75%, wherein the free area is defined
2 as the ratio of the free area between successive coil
3 turns and the cylinder that bisects the helical coil
4 circle.

5 In Figs. 1, 3 and 4, a gas conditioning
6 system 101 and fuel cells 100 to receive hydrogen are
7 in operative communication with reactor outlets 3.

8 Fig. 5 depicts yet another embodiment of the
9 present invention. In this embodiment shown
10 schematically the reaction chamber 116 is defined by
11 the annular space between an outer conduit 131 and an
12 inner conduit 132. The reactant gases enter the
13 reaction chamber through inlet means 112, and pass
14 through catalyst bed at 116 and then to space 134 at
15 the inlet of the inner conduit 132. The reactant gases
16 exit the inner conduit space through exit means 113.
17 The reactant gases passing through the inner conduit
18 132 transfer heat to the reactant gases contained in
19 the reaction chamber 116 to beneficially recuperate
20 heat from the endothermic reaction.

21 An axially extending radiant burner 107 is
22 vertically disposed within a combustion chamber 104.
23 The radiant burner is oriented in parallel with the
24 longitudinal extent of the tubular reaction conduit.
25 If a multiplicity of such tubular reaction conduits are
26 used, they can be oriented concentrically around a

1 centrally disposed radiant burner that uniformly
2 radiates in a 360 degree arc. The radiant burner
3 transfers radiant energy to the surface of the outer
4 conduits 131.

5 Combustion gases exiting the radiant burner
6 107 are introduced into a convection chamber 117 that
7 is concentrically disposed around a portion of the
8 outer conduit 131 in the proximity of the tubular
9 conduit end containing the reactant gas inlet means
10 112. After transferring heat by convection to the
11 outer conduit, the combustion gases exit at an outlet
12 means 111.

13 Accordingly, the Fig. 5 embodiment includes:

14 a) a straight tubular outer conduit
15 concentrically disposed around an inner conduit to form
16 a reaction chamber containing catalyst in the annular
17 space between the outer conduit wall and the inner
18 conduit wall, for conversion of hydrocarbon to
19 industrial gases by reaction with steam, and an inner
20 conduit defined space for the return flow of reactant
21 gases to an exit means; said tubular reaction chamber
22 having one end that extends into the combustion chamber
23 and an opposite end that extends outside of the
24 combustion chamber, and there being inlet means that is
25 in communication with the annular space and an exit

1 means that is in communication with the inner conduit
2 defined space,
3 b) and a radiant burner vertically disposed
4 within said combustion chamber and having a gas
5 permeable zone that promotes the flameless combustion
6 of fuel and oxidant supplied to said burner in order to
7 heat the metal fiber surface of the burner to
8 incandescence for radiating heat energy to the reaction
9 chamber.

10 Also, there is typically a convection chamber
11 extending about a portion of the tubular reaction
12 chamber in the proximity of the end containing the
13 reactant gas inlet and outlet means to enhance heat
14 transfer from combustion products; said convection
15 chamber having an inlet means that is in communication
16 with the combustion chamber and an exit means for
17 combustion products that is outside the combustion
18 chamber.

19 The structure may be alternatively considered
20 to represent a multiplicity of said tubular reaction
21 chambers are provided and are concentrically disposed
22 around a centrally located and vertically disposed
23 cylindrical radiant burner having a 360 degree radiant
24 arc.

1 It should be apparent to those skilled in the
2 art that the subject invention accomplishes the objects
3 set forth above.

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